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Sub-Poissonian light generation in the non-degenerate three-wave mixing is studied numerically and analytically within quantum and classical approaches. Husimi  $Q$ -functions and their classical trajectory simulations are analysed to reveal a special regime corresponding to the time-stable sub-Poissonian photocount statistics of the sum-frequency mode. Conditions for observation of this regime are discussed. Theoretical predictions of the Fano factor and explanation of the extraordinary stabilization of the sub-Poissonian photocount behavior are obtained analytically by applying the classical trajectories. Scaling laws for the maximum sub-Poissonian behavior are found. Noise suppression levels in the non-degenerate vs degenerate three-wave mixing are discussed on different time scales compared to the revival times. It is shown that the non-degenerate conversion offers much better stabilization of the suppressed noise in comparison to that of degenerate process.

**Keywords:** sub-Poissonian statistics, three-wave mixing, Fano factor, classical trajectories

## I. INTRODUCTION

For almost four decades, since the pioneering experiments of Franken et al. [1] and theoretical foundations laid down by Bloembergen et al. [2], multiwave mixing has unceasingly been in the forefront of quantum-optical investigations [3,4]. In particular, the *three-wave mixing* (TWM) has attracted considerable interest as a parametric non-linear process of conversion of two sub-frequency (say,  $\omega_1$  and  $\omega_2$ ) photons into one sum-frequency ( $\omega_1 + \omega_2 \rightarrow \omega_3$ ) photon, together with the inverse process. TWM can be observed in non-linear crystals like ADP, KDP, LiNbO<sub>3</sub> or BaTiO<sub>3</sub> [5]. Both the total energy,  $\hbar\omega_1 + \hbar\omega_2 = \hbar\omega_3$ , and momentum,  $\hbar\mathbf{k}_1 + \hbar\mathbf{k}_2 = \hbar\mathbf{k}_3$ , of interacting photons are conserved. TWM is observable for proper orientations of light beam polarizations and crystal axes [4], therefore it can be considered as a parametric process. TWM is used for the frequency-up conversion if  $\omega_1 \rightarrow \omega_3$  or the frequency-down conversion if  $\omega_3 \rightarrow \omega_1$ . The process is also useful for generation of non-classical light such as squeezed, sub-Poissonian and antibunched light [6].

Before the computer era, the quantum dynamics was usually investigated under the short-time approximation only. Nowadays, the Taylor series of quantum operators can be found for almost any number of terms with the help of fast computers and sophisticated software. However, these series are usually convergent for short evolution times or even for initial time only. Thus, numerical quantum methods (see, e.g., [7]) fail in simulation of the long-time quantum evolution. On the other hand, as we have shown in [8,9], the method of classical trajectories gives very good estimation in the case of strong-field

interaction (practically, for photon numbers larger than 10). The computational speed of the classical-trajectory method does not depend on the numbers of interacting photons and, moreover, for a larger number of photons one obtains better precision. Thus, the method is very fast and offers a simple substitute for the tedious exact quantum numerical calculations. The classical-trajectory method enables not only numerical but also some analytical predictions, e.g., for stationary Fano factors [8,9] or for maximum pump depletion in TWM [10]. A method similar to ours to simulate classical noise in TWM was used by Chmela [11].

In the previous papers, we have studied degenerate processes of wave mixing, including the second [8] and higher [9] harmonic generations. Here, we generalize the former results for the non-degenerate wave mixing. It is well-known that both degenerate and non-degenerate TWM can be used for generation of sub-Poissonian light [12,13]. Nevertheless, theoretical predictions of quantum parameters, like Fano factor, are most often derived under the short-interaction (short-time or short-length) approximation (see, e.g., [12–14]), thus valid for weak non-linear coupling of the optical fields only. Motivated by papers of Nikitin and Masalov [15] and of Bandilla, Drobný and Jex [16,17], we analyse the long-interaction evolution of TWM. The main result of this article can be summarized as follows: the TWM can be a source of time-stable sub-Poissonian light of the sum-frequency mode in the no-energy-transfer regime. The deepest noise reduction, with the Fano factor equal to  $5/6$ , can be observed for the balanced input amplitudes  $r_1 = r_2 = r_3/\sqrt{2}$ . The same degree of photocount noise suppression in the sum-frequency mode can be achieved for the degenerate TWM. However, the sub-Poissonian

light produced in non-degenerate TWM is much better stabilized compared to that in degenerate TWM. Moreover, the Fano factors for the sub-frequency modes in the non-degenerate TWM are smaller than those for the degenerate process. This and other results will be demonstrated analytically by applying a method of classical trajectories and tested numerically within quantum approach.

## II. QUANTUM ANALYSIS

In quantum approach, the non-degenerate three-wave mixing can be described by the interaction Hamiltonian (e.g., [12])

$$\hat{H} = \hbar g (\hat{a}_1 \hat{a}_2 \hat{a}_3^\dagger + \hat{a}_1^\dagger \hat{a}_2^\dagger \hat{a}_3), \quad (1)$$

where  $\hat{a}_k$  and  $\hat{a}_k^\dagger$  denote, respectively, annihilation and creation operators of the sub-frequency (labeled with subscripts 1, 2) and sum-frequency (subscript 3) modes;  $g$  is a non-linear coupling parameter, which is related to the quadratic susceptibility tensor  $\chi^{(2)}$  of a given non-linear optical crystal and also dependent on the geometry of laboratory set-up [4].

As in [9], we analyse the quantum Fano factors given by  $F_k = (\langle \hat{n}_k^2 \rangle - \langle \hat{n}_k \rangle^2) / \langle \hat{n}_k \rangle$  for a photon-number operator  $\hat{n}_k = \hat{a}_k^\dagger \hat{a}_k$ . The light is referred to as sub-Poissonian if  $F_k < 1$  and super-Poissonian if  $F_k > 1$ .

For weak non-linear interactions or short crystal, the short-time approximation can be applied for analytical predictions of photocount noise suppression with  $F_k < 1$  [12,13]. The Fano factors are approximated by

$$F_{1,2} = 1 + 2r_3^2 (gt)^2 + \frac{8}{3} r_1 r_2 r_3 \sin \theta (gt)^3 + \mathcal{O}\{(gt)^4\},$$

$$F_3 = 1 - \frac{4}{3} r_1 r_2 r_3 \sin \theta (gt)^3 + \mathcal{O}\{(gt)^4\}, \quad (2)$$

where  $r_k$  are the input coherent amplitudes and  $\theta = \phi_1 + \phi_2 - \phi_3$  is the input phase mismatch. For  $\sin \theta > 0$ , the sub-Poissonian statistics in the sum-frequency mode can be observed. For  $\theta = 0$ , we find the higher-order short-time Fano factor expansion to be

$$F_3 = 1 + (r_3^2 - 7r_1^2 r_2^2 + 4r_1^2 r_3^2 + 4r_2^2 r_3^2) \frac{(gt)^4}{3} + \mathcal{O}\{(gt)^5\}.$$

It is seen that the sub-Poissonian light in the sum-frequency mode is generated for some combinations of input amplitudes  $r_k$ . Since the Fano factors depend weakly on time (i.e., in its third or higher-order power), it is difficult to observe the sub-Poissonian light generation in the short-time regime.

To analyse the exact quantum dynamics of the TWM process beyond the short-time approximation, we apply

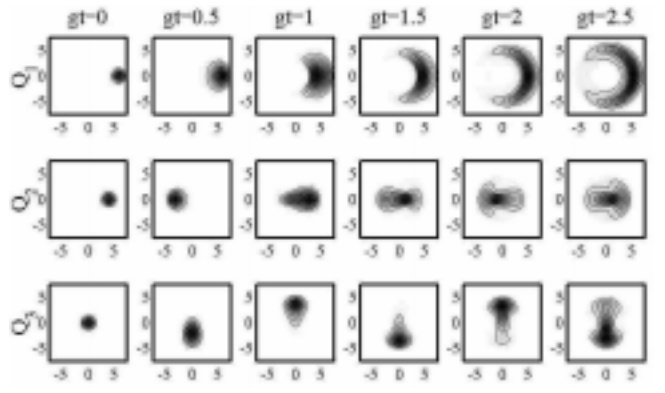


FIG. 1. Quantum dynamics *out* of NETR: Cross-sections of single-mode  $Q$ -functions:  $Q_1(\text{Re } \alpha_1, \text{Im } \alpha_1)$  and  $Q_2(\text{Re } \alpha_2, \text{Im } \alpha_2)$  for sub-frequency modes and  $Q_3(\text{Re } \alpha_3, \text{Im } \alpha_3)$  for sum-frequency mode at different scaled evolution times for initial coherent fields with real amplitudes,  $\alpha_k(0) = r_k$ , set to  $r_1 = 6$ ,  $r_2 = 4$  and  $r_3 = 0$ .

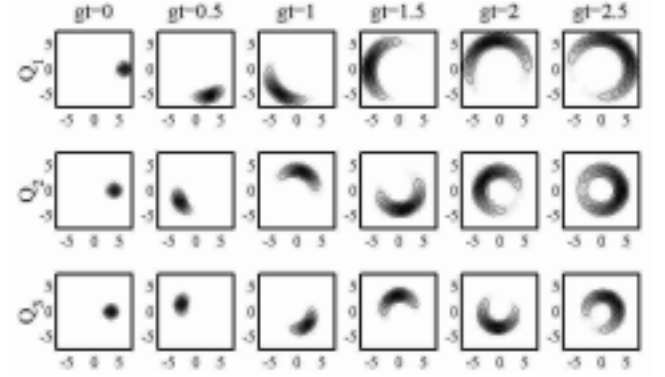


FIG. 2. Quantum dynamics *in* NETR: Cross-sections of marginal  $Q$ -functions as in figure 1, but for  $r_1 = 6$ ,  $r_2 = 4$ , and  $r_3 = r_1 r_2 / \sqrt{r_1^2 + r_2^2} \approx 3.328$ .

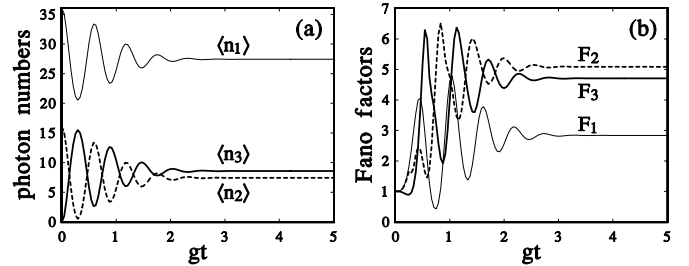


FIG. 3. Quantum dynamics *out* of NETR: (a) photon numbers  $\langle \hat{n}_k \rangle$  and (b) Fano factors  $F_k$  ( $k = 1, 2, 3$ ) for coherent inputs with real amplitudes  $r_1 = 6$ ,  $r_2 = 4$ , and  $r_3 = 0$ . For longer times, all the modes should be stationary with the super-Poissonian statistics,  $F_k > 1$ .

the Walls-Barakat method [3] of Hamiltonian diagonalization for the initial coherent states. Quantum analysis enables numerical estimation of all statistical properties including photocount noise. Complete quantum information of the TWM dynamics can be given by the Husimi

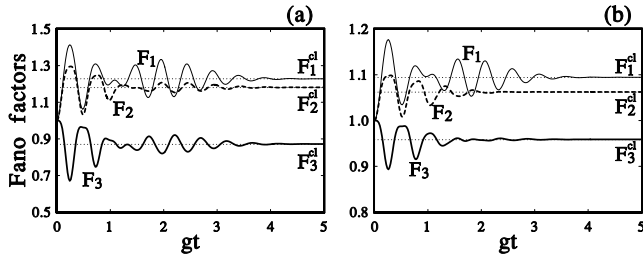


FIG. 4. Quantum dynamics *in* NETR: Fano factors  $F_1$  (thin solid),  $F_2$  (dashed), and  $F_3$  (thick solid curves) versus scaled time for different amplitudes of initial coherent fields: (a)  $r_1 = 6$ ,  $r_2 = 4$ , and (b)  $r_1 = 6$ ,  $r_2 = 2$  together with  $r_3 = r_1 r_2 / \sqrt{r_1^2 + r_2^2}$ . Sub-frequency modes become super-Poissonian,  $F_{1,2} > 1$ . By contrast, the sum-frequency mode becomes sub-Poissonian,  $F_3 < 1$ . Dotted lines represent the classical trajectory predictions of  $F_k^{\text{cl}}$  to which all the quantum curves tend asymptotically.

$Q$ -function defined to be

$$Q(\alpha_1, \alpha_2, \alpha_3) = \pi^{-3} \langle \alpha_1, \alpha_2, \alpha_3 | \hat{\rho} | \alpha_1, \alpha_2, \alpha_3 \rangle, \quad (3)$$

where  $|\alpha_1\rangle \otimes |\alpha_2\rangle \otimes |\alpha_3\rangle$ . In figures 1 and 2, we plot its marginal single-mode Husimi  $Q$ -functions given by

$$Q(\alpha_k) = \int Q(\alpha_1, \alpha_2, \alpha_3) \prod_{m \neq k} d^2 \alpha_m, \quad (4)$$

where  $k, m = 1, 2, 3$ . The Fano factors, presented in figures 3 and 4, were calculated with the help of the marginal  $Q$ -functions. Due to obvious computational difficulties, the exact quantum results can be obtained for relatively small numbers (up to few hundreds) of interacting photons only.

By analysing the numerical quantum solution we observe that the basic features of the photon number evolution for the non-degenerate TWM are in agreement with those for the harmonic generation processes, as recently reported in [8,9]. In particular, we observe the so-called no-energy-transfer regime (NETR) [18,16,17], for which the energies and intensities of both modes remain constant in time during the interaction. Although small energy flows between the modes appear as a consequence of vacuum fluctuations, their influence is negligible for strong fields. NETR in the three-wave mixing can be observed if the amplitudes and phases of the initial coherent fields are matched as follows (Eq. (18) in [16]):

$$\frac{1}{r_3^2} = \frac{1}{r_1^2} + \frac{1}{r_2^2}, \quad (5)$$

$$\phi_3 = \phi_1 + \phi_2.$$

Usually, i.e., for the initial coherent fields *not* satisfying (5), all the Fano factors are stabilized in the super-Poissonian statistics after a short ( $gt |r_k| \lesssim 1$ ) relaxation period. Thus, the outputs have high-level photocount noise. In figure 1, we present a typical quantum evolution

of the single-mode Husimi functions  $Q(\alpha_1)$  and  $Q(\alpha_2)$  for the initial amplitudes  $r_1 = 6$ ,  $r_2 = 4$  and  $r_3 = 0$ . The corresponding evolutions of the photon numbers and Fano factors are presented in figure 3.

Different behaviour is observed if the initial phase  $\phi_3$  and amplitude  $r_3$  of the sum-frequency mode fulfill the condition for NETR. This distinction is clearly seen by comparing figures 1 and 2 for  $Q$ -functions or figures 3 and 4 for the Fano factors. In figure 4, the Fano factors are calculated for two different pairs of the initial amplitudes of sub-frequency modes: (a)  $r_1 = 6$ ,  $r_2 = 4$  and (b)  $r_1 = 6$ ,  $r_2 = 2$  and the sum-frequency-mode amplitude  $r_3$  fulfilling (5). We observe that all the Fano factor curves start at  $F_k(0) = 1$  and after some relaxations become stationary at much lower noise levels than those for fields

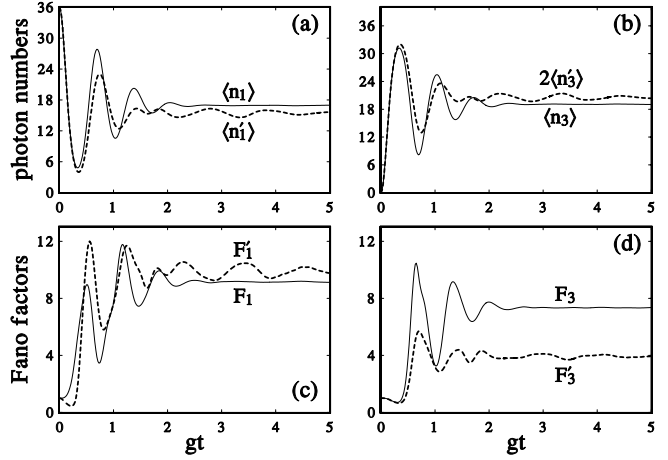


FIG. 5. Non-degenerate vs degenerate TWM *out* of NETR: Photon numbers  $\langle \hat{n}_k \rangle$  and Fano factors  $F_k$  ( $k = 1, 3$ ) are obtained for initial amplitudes  $r_1 = r_2 = 6$  and  $r_3 = 0$ , whereas  $\langle \hat{n}'_k \rangle$  and  $F'_k$  are for  $r'_1 = 6$  and  $r'_3 = 0$ . All quantities with (without) prime correspond to the degenerate (non-degenerate) TWM.

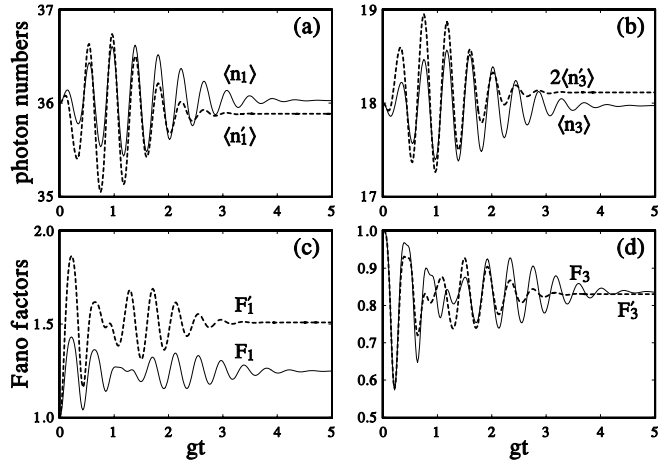


FIG. 6. Non-degenerate vs degenerate TWM *in* NETR:  $\langle \hat{n}_k \rangle$  and  $F_k$  are calculated for initial amplitudes  $r_1 = r_2 = 6$  and  $r_3 = 6/\sqrt{2}$ , whereas  $\langle \hat{n}'_k \rangle$  and  $F'_k$  are for  $r'_1 = 6$  and  $r'_3 = 3$ .

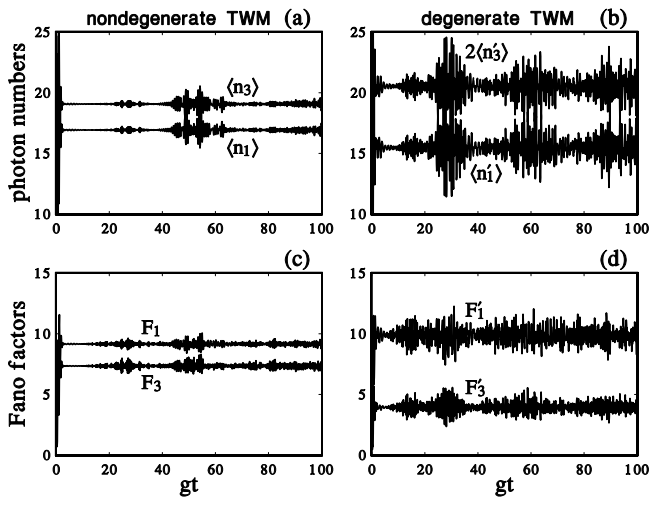


FIG. 7. Revivals and collapses in degenerate TWM vs those in non-degenerate TWM *out of* NETR. Initial conditions are the same as in figure 5.

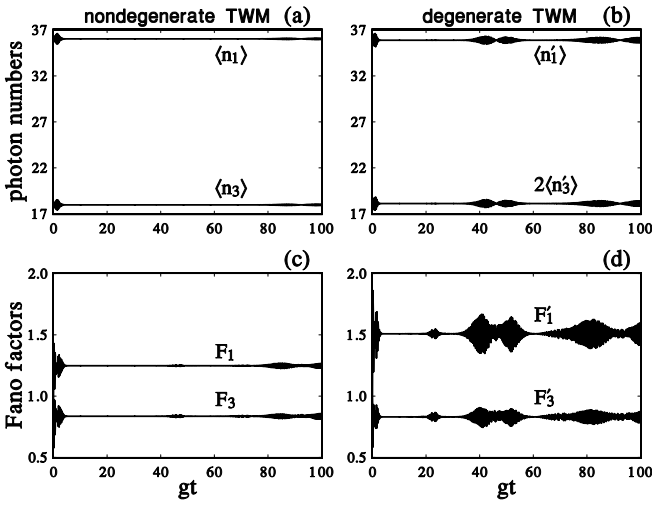


FIG. 8. Revivals and collapses in degenerate and non-degenerate TWM *in* NETR with the same initial conditions as those in figure 6.

out of NETR. The sub-frequency modes remain super-Poissonian with  $F_{1,2}(t) > 1$ , whereas the sum-frequency mode becomes sub-Poissonian with  $F_3(t) < 1$ . The most suppressed noise is observed for the balanced inputs, given by  $r_1 = r_2$  and  $r_3 = r_1/\sqrt{2}$ , as a special case of condition (5). For those inputs, the Fano factor in the time limit tends to  $F_3(t \rightarrow \infty) \approx 0.83$ .

As we have shown in Refs. [8,9], the same degree of the Fano factor can be obtained in degenerate TWM. Thus, one can address the following objection: What is the point to study the same parameter in the closely related non-degenerate version? First, we stress that the same Fano factor is obtained in a special case only: for the sum-frequency mode in NETR for long-interaction times and high-intensity fields. By contrast, these factors are distinct for the sub-frequency modes under the

same initial conditions. In general, the results even for the sum-frequency mode in these two processes are different for initial conditions either not fulfilling (5) or for lower intensity fields or different time periods. Second, we will show that the Fano factors for non-degenerate TWM are better stabilized than those for degenerate TWM for much longer evolution times. This is an important advantage of the non-degenerate conversion.

For better comparison, let us analyse in detail the degenerate TWM described by

$$\hat{H}' = \hbar g' (\hat{a}_1^2 \hat{a}_3^\dagger + \hat{a}_1^{\dagger 2} \hat{a}_3), \quad (6)$$

For clarity, observables calculated for the degenerate TWM are marked with prime to distinguish them from those for model (1) and we keep subscript ‘3’ (not ‘2’) for the sum-frequency mode. Hamiltonian (6) formally differs from (1) in the assumption of  $\hat{a}_1 = \hat{a}_2$  only. But we also put  $g' = g/\sqrt{2}$  for better synchronization of oscillations in photon numbers  $\langle \hat{n}_k' \rangle$  and  $\langle \hat{n}_k \rangle$ . Sub-Poissonian statistics in degenerate TWM was analysed in our former works [8,9]. In figures 5 and 6, we compare exact quantum evolutions of the mean photon numbers and Fano factors for degenerate ( $\langle \hat{n}_k' \rangle$  and  $F_k'$  for  $k = 1, 3$ ) and non-degenerate ( $\langle \hat{n}_k \rangle$  and  $F_k$ ) TWM for the same initial conditions. We observe similar behavior for short times  $gt \ll 1$  only. For longer times (also for  $gt < 1$ ) results are not equal by no means. Different predictions of quantum evolutions for models (1) and (6) come from different commutation relations:  $[\hat{a}_1, \hat{a}_2^\dagger] = 0$  for non-degenerate TWM and  $[\hat{a}_1, \hat{a}_2^\dagger] \equiv [\hat{a}_1, \hat{a}_1^\dagger] = 1$  for degenerate TWM. As a result, constants of motion are different:  $\hat{N}'_{\text{total}}(t) \equiv \hat{n}_1'(t) + 2\hat{n}_3'(t) = \text{const}$  for degenerate TWM, whereas  $\hat{n}_1(t) - \hat{n}_2(t) = \text{const}$  and  $\hat{n}_1(t) + \hat{n}_3(t) = \text{const}$  for non-degenerate TWM. We note that  $\hat{N}_{\text{total}}(t) \equiv \hat{n}_1(t) + \hat{n}_2(t) + \hat{n}_3(t) \neq \text{const}$  in the latter process. Evolutions, presented in figures 5 and 6 for degenerate and non-degenerate TWM, are distinct in amplitudes and frequency of oscillations as well as in the level of their “saturation”. For example, the limiting value of the sum-frequency Fano factor for degenerate TWM is lower than that for non-degenerate TWM for evolution out of NETR (see figure 5(d)). While the sub-frequency Fano factor in NETR is considerably higher for degenerate compared to non-degenerate TWM (see figure 6(c)).

In figures 1–6, we have analysed the time regime which is long compared to the typical interaction times for known crystal lengths. However, the time is short compared to the revival times for such systems. The question arises about the photon-number noise suppression on such a long scale. This analysis will show an advantage of non-degenerate over degenerate TWM related to the stabilization of the suppressed photon-number noise. In figures 7 and 8, we present the long-time evolution for  $0 < gt < 100$  of the mean photon numbers and Fano factors for both non-degenerate ( $\langle \hat{n}_k \rangle$  and  $F_k$ ) and degenerate ( $\langle \hat{n}_k' \rangle$  and  $F_k'$ ) TWM. We observe that the revivals

are strongly pronounced for (i) degenerate TWM (right figures 7 and 8) compared to non-degenerate process (left figures), and (ii) outside NETR (figure 7) rather than in NETR (figure 8). Thus, the non-degenerate TWM in NETR exhibits the highest stability. Even for longer evolution times as  $100 < gt < 1000$  of non-degenerate TWM, the oscillations are similar to those for  $0 < gt < 100$  and it is hardly difficult to classify them as a typical revival. Oscillations in  $\langle \hat{n}_k \rangle$  are of order  $10^{-3}$  and in  $F_k$  are of order  $10^{-2}$  even for such small initial amplitudes equal to  $\alpha_1 = \alpha_2 = 6$  and  $\alpha_3 = 6/\sqrt{2}$ . Our analysis is restricted to initial coherent inputs. It is worth noting that the revivals are much stronger for quantum input fields like, e.g., Fock states. In conclusion, although the degenerate and non-degenerate TWM lead to approximately the same photon-number noise suppression in the sum-frequency mode for NETR (see figure 6(d)), the non-degenerate process offers much better stabilization of the suppressed noise for long evolution times (compare figures 8(c) and 8(d)).

In the next sections, we will apply an approximate method of classical trajectories to explain the extraordinary numerical stabilization of the observed photocount noise and to estimate analytically the level of noise suppression for NETR.

### III. CLASSICAL ANALYSIS

Complete quantum solution of the model given by Hamiltonian (1) can be found numerically only. Yet, in a special case for strong fields, analytical results can be obtained by applying approximate classical methods.

In analogy to quantum Hamiltonian (1), the classical model of non-degenerate TWM can be described by [4]:

$$\mathcal{H} = g(\alpha_1 \alpha_2 \alpha_3^* + \text{c.c.}), \quad (7)$$

where  $\alpha_k$  are the complex amplitudes of the  $k$ -th mode and  $g$  is a non-linear coupling parameter. From (7), one readily obtains the following complex differential equations

$$\begin{aligned} \dot{\alpha}_1 &= -ig\alpha_2^* \alpha_3, \\ \dot{\alpha}_2 &= -ig\alpha_1^* \alpha_3, \\ \dot{\alpha}_3 &= -ig\alpha_1 \alpha_2. \end{aligned} \quad (8)$$

It is easy to show by comparing (8) with equations (12) and (13) from Ref. [8] that the classical models for the degenerate and non-degenerate TWM are equivalent for  $\alpha_1 = \alpha_2$  and arbitrary evolution times. To get equations of motion for degenerate TWM, it is enough to replace  $g$  by  $\sqrt{2}g'$  and  $\alpha_3$  by  $\sqrt{2}\alpha'_3$  in (8). As was discussed in the former section, the quantum evolutions of degenerate and non-degenerate TWM are equivalent for  $gt \ll 1$  only.

By introducing real amplitudes and phases,  $\alpha_k = r_k e^{i\phi_k}$ , equations (8) can be transformed into the following four real equations

$$\begin{aligned} \dot{r}_1 &= -gr_2 r_3 \sin \theta, \\ \dot{r}_2 &= -gr_1 r_3 \sin \theta, \\ \dot{r}_3 &= gr_1 r_2 \sin \theta, \\ \dot{\theta} &= g \left( \frac{r_1 r_2}{r_3} - \frac{r_1 r_3}{r_2} - \frac{r_2 r_3}{r_1} \right) \cos \theta, \end{aligned} \quad (9)$$

where  $\theta = \phi_1 + \phi_2 - \phi_3$  is the phase mismatch. The system (9) has three integrals of motion

$$\begin{aligned} E_1 &= r_1^2 + r_3^2 = n_1 + n_3, \\ E_2 &= r_2^2 + r_3^2 = n_2 + n_3, \\ K &= r_1 r_2 r_3 \cos \theta. \end{aligned} \quad (10)$$

By extracting  $r_1, r_2$  and  $\theta$  from (9), equation for the remaining amplitude  $r_3$  reads as

$$(r_3 \dot{r}_3 / g)^2 + K^2 = r_3^2 (E_1 - r_3^2) (E_2 - r_3^2) \quad (11)$$

or, equivalently, for the intensity  $n_3 = r_3^2$  as

$$\begin{aligned} (\dot{n}_3 / 2g)^2 &= n_3 (E_1 - n_3) (E_2 - n_3) - K^2 \\ &= (a - n_3) (b - n_3) (n_3 - c), \end{aligned} \quad (12)$$

where the numbers  $a \geq b \geq c$  are the roots of cubic equation  $n_3 (E_1 - n_3) (E_2 - n_3) - K^2 = 0$  satisfying the conditions  $abc = K^2$ ,  $a + b + c = E_1 + E_2$ , and  $ab + ac + bc = E_1 E_2$ . Then, the solution for  $n_3(t)$  is found to be

$$n_3(t) = c + (b - c) \text{sn}^2 [\sqrt{a - c}gt + \phi_0, k], \quad (13)$$

where  $\text{sn}(u, k)$  is the Jacobi elliptic function with  $k = \sqrt{\frac{b-c}{a-c}}$  and  $\phi_0$  is the initial phase given by the elliptic integral of the first kind

$$\phi_0 = F(z, k) = \int_0^z \frac{dx}{\sqrt{1 - k^2 \sin^2 x}}, \quad (14)$$

where  $z = \arcsin \sqrt{(n_3(0) - c)/(b - c)}$ . One observes that  $n_3$  is a periodic function oscillating between the values  $c$  and  $b$  with the period given by  $T = 4F(\frac{\pi}{2}, k)/g$ .

In two special cases, solution (13) reduces to the elementary solutions:

$$n_3(t) = r^2 \tanh^2(rgt) \quad (15)$$

for  $r_1 = r_2 = r$  and  $r_3 = 0$ , and

$$n_3(t) = r^2 \text{sech}^2(rgt) \quad (16)$$

for  $r_1 = r_2 = 0$  and  $r_3 = r$ . Another elementary solution is obtained for the initial fields fulfilling conditions (5). In this case, the solution reads as ( $k = 1, 2, 3$ )

$$\alpha_k(t) = r_k \exp \left( -i \frac{r_1 r_2 r_3}{r_k^2} gt \right), \quad (17)$$

which describes the classical *no-energy-transfer regime* (NETR) [18], since the amplitudes and energies of all interacting modes remain constant, i.e.,  $n_k(t) = |\alpha_k(t)|^2 = r_k^2$ . We conclude that NETR observed in the quantum numerical analysis presented in former section corresponds to the classical solution (17).

Classical solutions, as presented in the preceding section, do not describe quantum noise. Nevertheless, they can be used for simulation of quantum noise if the initial complex amplitudes are chosen randomly. This approach, referred to as the method of *classical trajectories*, has been applied successfully in a description of noise in various quantum-optical phenomena [8–11, 15–20]. By analysing  $Q$ -functions and Fano factors, we will show that the method of classical trajectories properly simulates photocount noise in the TWM processes.

To calculate statistical moments, like the Fano factors, one needs to analyse the classical evolution of each process (trajectory) separately and then to average the moments over all the obtained trajectories. The classical Fano factor, defined to be

$$F^{\text{cl}} = \frac{\overline{n^2} - \bar{n}^2}{\bar{n}}, \quad (18)$$

can be obtained by the classical trajectory averaging. We denote this averaging by bar to distinguish it from quantum ensemble averaging indicated by brackets. We will apply the method of classical trajectories along the lines of the analysis presented in [8]. We choose the initial amplitudes to be  $\alpha_k = r_k$  and blur them with the Gaussian noise, which results in

$$\alpha_k = r_k + x_k + iy_k, \quad (19)$$

where  $x_k$  and  $y_k$  are mutually independent real Gaussian stochastic quantities with the identical variances  $\sigma^2 = 1/4$ . We assume further that the fields are strong, i.e.,  $r_k \gg 1$ .

By substituting (19) into (10), the integrals of motion can be expressed as

$$\begin{aligned} E_1 &= |\alpha_1|^2 + |\alpha_3|^2 = r_1^2 + r_3^2 + c_1, \\ E_2 &= |\alpha_2|^2 + |\alpha_3|^2 = r_2^2 + r_3^2 + c_2, \\ K &= \text{Re}(\alpha_1 \alpha_2 \alpha_3^*) = r_1 r_2 r_3 + d_1, \end{aligned} \quad (20)$$

where  $c_1$ ,  $c_2$  and  $d_1$  are small corrections of  $E_1$ ,  $E_2$  and  $K$ , respectively, depending on  $r_k$ ,  $x_k$  and  $y_k$ . To eliminate the linear term in the RHS of (12), we substitute

$$\begin{aligned} n_3 &= \frac{1}{3}E_2 + \frac{1}{3}E_1 - \frac{1}{3}\sqrt{(E_2^2 - E_1 E_2 + E_1^2)} + \epsilon \\ &= n_{30} + b + \epsilon, \end{aligned} \quad (21)$$

where  $\epsilon$  is a small correction of stationary value  $n_{30} + b$ . Under the strong-field approximation ( $r_k \gg 1$ ), one can neglect the small correction  $\epsilon^3$  and the RHS of (12) can be approximated by the quadratic function  $\Omega^2(a^2 - \epsilon^2)$ . Thus, one gets simple equation

$$\dot{\epsilon}^2 = (2g\Omega)^2(a^2 - \epsilon^2), \quad (22)$$

which leads to the solution of (12) in the following form

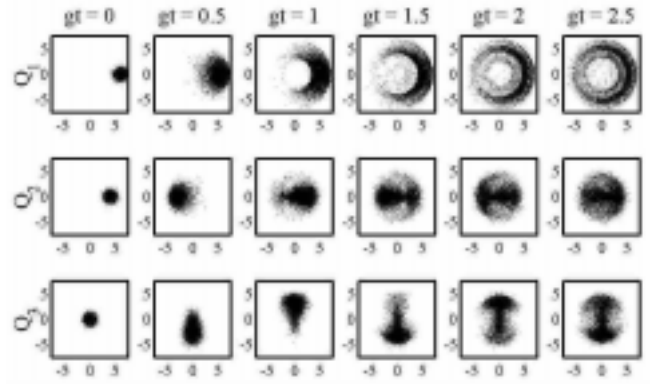


FIG. 9. Classical simulation of typical quantum dynamics *out* of NETR: Clouds of 10,000 points representing marginal  $Q$ -functions for the same initial conditions and times as in figure 1.

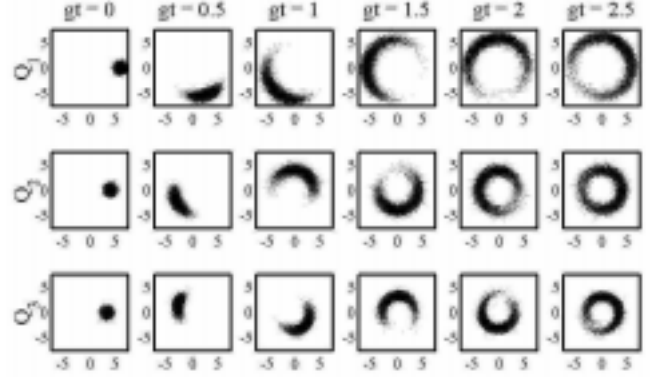


FIG. 10. Classical simulation of quantum dynamics *in* NETR for the same cases as in figure 2.

$$n_3(t) = n_{30} + b + a \sin(2g\Omega t + \varphi), \quad (23)$$

where  $n_{30} = r_3^2 = r_1^2 r_2^2 / (r_1^2 + r_2^2)$ . The coefficients  $a$ ,  $b$  and  $\Omega$ , together with  $c_1$  and  $c_2$ , are complex functions of  $r_k$  and noise parameters  $x_k$  and  $y_k$ . With the help of integrals of motions, given by (10), solutions for other modes ( $k = 1, 2$ ) can readily be found as

$$n_k(t) = E_k - n_3(t) = r_k^2 + c_k - b - a \sin(2g\Omega t + \varphi). \quad (24)$$

We observe that all the three solutions, given by (23) and (24), are of the form of large constants slightly perturbed by the same harmonic function. In figures 9 and 10, we present classical simulation of quantum dynamics by calculating time evolutions of 10,000 points in phase space according to classical equations of motion. These representations correspond to the Husimi  $Q$ -functions presented in figures 1 and 2, respectively. By comparing figures 1 and 2 or, equivalently, figures 9 and 10, we observe two distinct types of evolution determined by the initial amplitudes to be in NETR or out of it.

The classical and quantum descriptions are principally different in detail. Thus, our plots of the  $Q$ -function

based on the exact quantum solution of TWM (figures 1 and 2) and those obtained by an approximate classical simulation (figures 9 and 10) also differ in detail. The discrepancies are more pronounced for lower amplitude inputs and longer interactions. Moreover, the methods of graphical representations are different: a topographical picture of  $Q_k(\alpha)$  versus a cloud of classical points. Nevertheless, “it is surprising how close the clouds of dots are to the  $Q$ -function” [15]. The clear correspondence between figures 2 and 10 or 1 and 9 justifies our application of the classical trajectory approximation.

As the next step of the classical trajectory method, one has to perform averaging of solutions (23) and (24) to calculate the desired moments. We find that the mean values of the parameters occurring in solution (24) are  $\bar{b} = \bar{c}_1 = \bar{c}_2 = 0$  and

$$\bar{\Omega} = \sqrt{r_1^2 + r_2^2 - r_3^2} = \sqrt{\frac{r_2^4 + r_1^2 r_2^2 + r_1^4}{r_1^2 + r_2^2}} \quad (25)$$

together with their mean quadratic moments

$$\begin{aligned} \overline{b^2} &= \frac{2A}{r_2^2 + r_1^2} (2r_1^8 + r_1^6 r_2^2 + 2r_1^4 r_2^4 + r_1^2 r_2^6 + 2r_2^8), \\ \overline{(c_1 - b)^2} &= \frac{2A}{r_2^2} (4r_1^8 + 10r_1^6 r_2^2 + 11r_1^4 r_2^4 + 7r_1^2 r_2^6 + 2r_2^8), \\ \overline{(c_2 - b)^2} &= \frac{2A}{r_2^2} (2r_1^8 + 7r_1^6 r_2^2 + 11r_1^4 r_2^4 + 10r_1^2 r_2^6 + 4r_2^8), \\ \overline{a^2} &= 2A(4r_1^6 + 7r_1^4 r_2^2 + 7r_1^2 r_2^4 + 4r_2^6), \end{aligned} \quad (26)$$

given in terms of the auxiliary function

$$A = \frac{r_1^2 r_2^2}{8(r_1^4 + r_1^2 r_2^2 + r_2^4)^2}. \quad (27)$$

The phase  $\varphi$  can be obtained from (23) at  $t = 0$ . Thus, the photon-number mean values are simply equal to  $\bar{n}_k = r_k^2$  and their variances are given by ( $k = 1, 2, 3$ )

$$\overline{n_k^2} - \bar{n}_k^2 = \overline{(c_k - b)^2} + \frac{1}{2} \overline{a^2} \quad (28)$$

in terms of the moments (26) and  $c_3 \equiv 0$ . The term  $\sin^2(2g\Omega t + \varphi)$  has simply been estimated as  $\frac{1}{2}$  for sufficiently large  $t$ , when  $\bar{n}_k$  and  $F_k$  become time-independent. Thus, we arrive at the following Fano factors

$$\begin{aligned} F_1^{\text{cl}} &= 1 + A(8r_1^4 + 5r_1^2 r_2^2 + 5r_2^4), \\ F_2^{\text{cl}} &= 1 + A(5r_1^4 + 5r_1^2 r_2^2 + 8r_2^4), \\ F_3^{\text{cl}} &= 1 - 3A(r_1^2 + r_2^2)^2, \end{aligned} \quad (29)$$

where  $A$  is given by (27). As one could expect, the formulas for  $F_k^{\text{cl}}$  are symmetric with respect to exchange of the subscripts  $1 \longleftrightarrow 2$ . We finally conclude that the TWM in the no-energy-transfer regime can be a source of time-stable sub-Poissonian light in the sum-frequency mode as described by the Fano factor

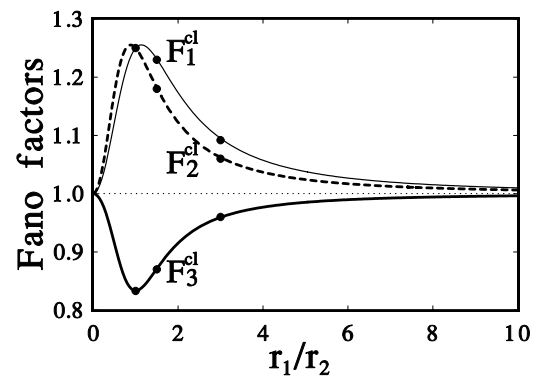


FIG. 11. Classical predictions of Fano factors  $F_k^{\text{cl}}$  ( $k = 1, 2, 3$ ) versus ratio  $r_1/r_2$  of the input coherent-field amplitudes. Small circles represent the quantum Fano factors  $F_k$  obtained from the exact quantum solutions presented in figures 4 and 6.

$$F_3^{\text{cl}}(\rho) = 1 - \frac{3\rho(1+\rho)^2}{8(1+\rho+\rho^2)^2} \leq 1, \quad (30)$$

depending on ratio of the mean intensities of initial coherent fields defined by  $\rho = r_1^2/r_2^2$  if  $r_1 > 0$  or  $\rho = r_2^2/r_1^2$  if  $r_2 > 0$ . The sub-frequency fields become super-Poissonian with the Fano factors

$$F_1^{\text{cl}}(\rho) = F_2^{\text{cl}}(1/\rho) = 1 + \frac{\rho(5+5\rho+8\rho^2)}{8(1+\rho+\rho^2)^2} \geq 1. \quad (31)$$

In figure 11, the classical predictions of the Fano factors are depicted as a function  $r_1/r_2$ . By analysing (30) and figures 4, 6, and 11, we conclude that the sum-frequency mode solely is sub-Poissonian ( $F_3^{\text{cl}} \leq 1$ ) and the strongest noise suppression is obtained for  $r_1 = r_2 = r_3/\sqrt{2}$ , when  $F_3^{\text{cl}} = 5/6 \approx 0.833$ . For highly unbalanced input intensities  $r_1 \ll r_2$  or  $r_1 \gg r_2$ , all the Fano factors approach unity  $F_1^{\text{cl}} \approx F_2^{\text{cl}} \approx F_3^{\text{cl}} \rightarrow 1$ . Mutually equal Fano factors, estimated by  $F_1^{\text{cl}} = F_2^{\text{cl}} = 5/4 = 1.25$ , are predicted for the balanced inputs  $r_1 = r_2 = r_3/\sqrt{2}$ . The maximum values of the Fano factors, estimated by  $F_1^{\text{cl}} = F_2^{\text{cl}} = \max = 1.255$ , are obtained for slightly unbalanced inputs:  $r_1 = 1.136r_2$  to maximize  $F_1^{\text{cl}}$  and for  $r_1 = 0.881r_2$  to maximize  $F_2^{\text{cl}}$ .

We have predicted in [8,9] the stationary sub-Poissonian Fano factors for the second ( $F_2^{\text{cl}} = 5/6$ ) and third ( $F_3^{\text{cl}} = 13/16$ ) harmonic generations within NETR. The minimum value of the sum-frequency-mode Fano factor for the non-degenerate TWM is the same as that obtained for degenerate TWM (i.e., second-harmonic generation) [8], but higher than that for degenerate four-wave mixing (i.e., third-harmonic generation). However, for the sub-frequency modes, the Fano factors for the non-degenerate TWM are smaller than those for the degenerate cases, namely  $F_{1,2}^{\text{cl}}(1) = 5/4$  instead of  $3/2$  and  $29/16$ , respectively.

In figures 3 and 4, we have compared evolutions of the exact quantum Fano factors  $F_k$  (depicted by solid or dashed curves) with their classical estimations,  $F_k^{\text{cl}}$

(dotted lines). One observes that all the curves start at  $F_k = 1$  for initial coherent fields and after some relaxations become quasi-stationary with  $F_k \approx F_k^{\text{cl}}$ , given by (29). It is worth noting that very good estimation is achieved even for relatively small amplitudes, e.g.,  $r_k \leq 6$ . We conclude that the conditions for NETR in quantum dynamics and suppression of the observed quantum noise levels are well explained by the classical trajectory method.

Finally, we will compare scaling properties of the Fano factors in their dependence on light intensity and initial amplitude for the degenerate and non-degenerate TWM. Drobný et al. [17] calculated the scaling laws under the truncated Wigner approximation for the maximum sub-Poissonian photon-number noise in TWM. Their formulas are valid also in the limit of  $r = \alpha_1(0) \rightarrow \infty$ . Here, we focus on nonlinear fits for finite ranges of  $r$  only. Let us investigate the scaling properties of the maximum sub-Poissonian character of the sum-frequency mode corresponding to the first minimum of the  $F_3$  and  $F'_3$  curves in figures 3(b), 4, 5(d), and 6(d). In figure 12, we plot the exact quantum numerical values of  $\min_t F_3(x, t) \equiv F_3(x, t_{\min})$  or  $\min_t F'_3(x, t) \equiv F'_3(x, t'_{\min})$  as a function of initial amplitudes  $x = r_1 = r_2 = r'_1$  and of intensities  $x \equiv \langle \hat{n}_3(t_{\min}) \rangle = \langle \hat{n}'_3(t'_{\min}) \rangle$ . We fit those minima with the exponent and polynomial functions of parameters listed in tables I and II. The errors, given in the last column, are estimated by the standard deviation. We observe that the scaling laws  $ax^b$  for non-degenerate

TABLE I. Scaling laws  $ax^b$  for non-degenerate TWM ( $\min_t F_3(x)$ ) and degenerate TWM ( $\min_t F'_3(x)$ ) *out* of NETR. Initial conditions are  $1 < r_1 = r_2 \leq 10$ ,  $1 < r'_1 \leq 20$ , and  $r_3 = r'_3 = 0$ .

No.	fitted function	$a$	$b$	error
1	$\min_t F_3(r_1)$	0.8819	-0.1254	0.0004
2	$\min_t F_3(\langle \hat{n}_3 \rangle)$	0.8560	-0.0572	0.0001
3	$\min_t F'_3(r'_1)$	0.7694	-0.0906	0.0003
4	$\min_t F'_3(\langle \hat{n}'_3 \rangle)$	0.7352	-0.0427	0.0003

TABLE II. Polynomial fit  $(ax^2 + bx + c)/x^2$  for non-degenerate ( $\min_t F_3(x)$ ) and degenerate ( $\min_t F'_3(x)$ ) TWM *in* NETR. Initial conditions are  $1 < r_1 = r_2 \leq 10$ ,  $1 < r'_1 \leq 20$ ,  $r_3 = r_1/\sqrt{2}$ , and  $r'_3 = r'_1/2$ .

No.	fitted function	$r_1$	$a$	$b$	$c$	error
1	$\min_t F_3(r_1)$	$> 1$	0.5474	0.1104	0.2956	0.0003
		$\gg 1$	0.5519	0.0643	0.3894	0.0007
2	$\min_t F_3(\langle \hat{n}_3 \rangle)$	$> 1$	0.5562	0.3143	-0.1618	0.0001
		$\gg 1$	0.5559	0.3240	-0.2057	0.0008
3	$\min_t F'_3(r'_1)$	$> 1$	0.5495	0.1195	0.3510	0.0003
		$\gg 1$	0.5541	0.0406	0.5739	0.0012
4	$\min_t F'_3(\langle \hat{n}'_3 \rangle)$	$> 1$	0.5558	0.2057	-0.0607	0.0001
		$\gg 1$	0.5556	0.2121	-0.0783	0.0005

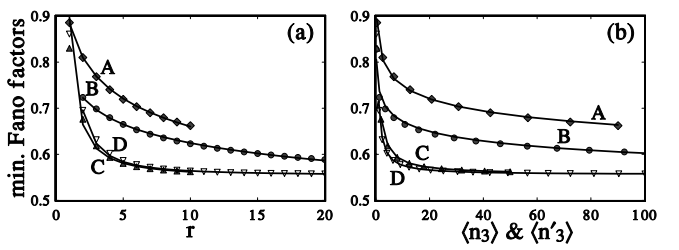


FIG. 12. Maximum photon-number noise suppression for non-degenerate (curves A and C) and degenerate (B and D) TWM: Time-minimized Fano factors,  $\min_t F_3$  and  $\min_t F'_3$ , as a function of (a) initial amplitudes  $r \equiv r_1 = r'_1$  and (b) mean photon numbers  $\langle \hat{n}_3 \rangle = \langle \hat{n}'_3 \rangle$ . Curves: A (with diamonds) for initial amplitude  $r_3 = 0$ , B (with circles) for  $r'_3 = 0$ , C (with solid triangles) for  $r_3 = r_1/\sqrt{2} = r/\sqrt{2}$ , D (with empty triangles) for  $r'_3 = r'_1/2 = r/2$ . The marked points are obtained from the exact quantum solutions and are fitted with the functions given in tables I and II.

and degenerate TWM give good approximation of the exact values *out* of NETR only. However, the  $ax^b$  law fails to describe with good precision  $\min_t F_3(x)$  and  $\min_t F'_3(x)$  *in* NETR at least for intensities up to 100 photons. Thus instead of the exponent law we apply the (inverse) polynomial fit of the form  $(ax^2 + bx + c)/x^2$ , where we introduce  $x^{-2}$  in relation to the definition of Fano factor. By contrast to the exponent fits, the polynomial laws give very good predictions of the maximum sub-Poissonian behavior at least for  $1 < r_1 = r_2 \leq 10$  and  $1 < r'_1 \leq 20$  as seen in figure 12 and table II. The scaling laws of Drobný et al. [17] differ slightly from ours presented in table I. The minor differences in the fitted parameters result from different ranges of  $r$  used in the fitting procedures and from application of the truncated Wigner approximation in Ref. [17] compared to our exact quantum method.

On the other hand, the Fano factors for the balanced ( $r_1 = r_2$ ) non-degenerate TWM under NETR conditions for long times and high intensity fields do not depend on light intensity, which follows from equations (30) and (31). Similarly, there are no scaling properties of the Fano factors in the degenerate TWM for long times and high intensity fields in NETR, i.e., under the same conditions as those assumed in our classical trajectory analysis.

## V. CONCLUSIONS

We have analysed the long-time interactions in the non-degenerate three-wave mixing. To the best of our knowledge, our quantum analysis is the first presentation of the exact and completely quantum solution of the nondegenerate three-wave mixing. In literature, a special solution can be found for initial sub-frequency fields with zero amplitudes  $\alpha_1 = \alpha_2 = 0$  only. The no-energy-transfer regime for proper choices of amplitudes and phases of the initial coherent fields has been



observed. We have compared the evolutions of the Husimi  $Q$ -functions and their classical trajectory simulations for processes in the no-energy-transfer regime and out of it. We have shown numerically in the quantum-mechanical approach that the three-wave mixing in the no-energy-transfer regime exhibits the time-stable photocount statistics. This phenomenon was explained analytically by applying the method of classical trajectories. We have shown that the sub-frequency modes become super-Poissonian with the Fano factor  $F_{1,2} > 1$ , whereas the sum-frequency mode becomes sub-Poissonian with  $F_3 < 1$ . We have found that the most suppressed photocount noise, given by  $F_3 \approx 5/6$ , is obtained for the balanced initial intensities  $r_1^2 = r_2^2$  of the sub-frequency modes and the sum-frequency intensity equal to  $r_3^2 = r_1^2/2$  as determined from condition (5) for the no-energy-transfer regime. Scaling laws and polynomial fits for the maximum sub-Poissonian behavior have been found for different processes and initial conditions. We have compared in detail the non-degenerate and degenerate conversions on time scales short and long compared to the revival times. We have observed that the non-degenerate three-wave mixing, contrary to the degenerate conversion, exhibits stabilization of the suppressed photon-number noise even on the revival time scale.

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